

Ultrafast and ultrasmall: focusing on atoms

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Wksp on Ultrafast X-ray Science 2004

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Collaborators

Atomic Physics Group

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X-ray Optics : *Eric Dufresne (MHATT-CAT)*

Ultrafast Laser: *David Reis, Matt DeCamp (U Michigan)
Eric Landahl (MHATT-CAT, APS)
Rob Crowell, Dave Gosztola (ANL)*

Outline

- **Motivation**
- **Background**
 - **threshold shift for valence photoionization**
- **First experiment**
 - **threshold shift for inner shell photoionization**
 - *technical challenges*
 - *experimental strategies*
 - *results so far*
- **Summary and outlook**

X-ray facilities: toward ultrafast & ultraintense

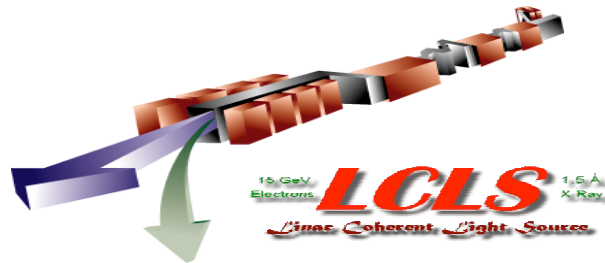


Single bunch specs

10^8 x-rays 87 ps



$\approx 10^7$ x-rays 80 fs

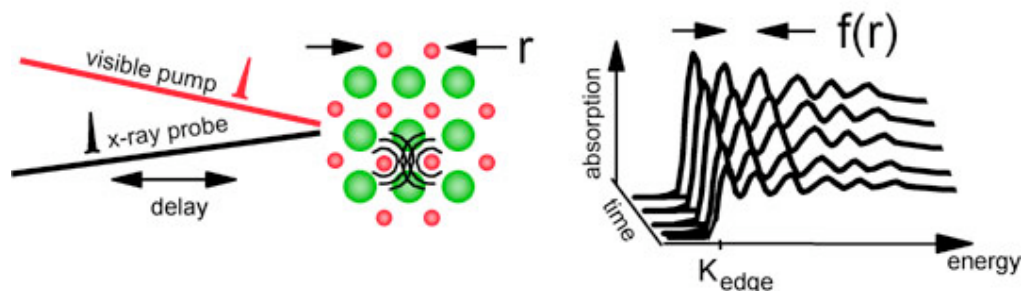


10^{12} x-rays 230 fs

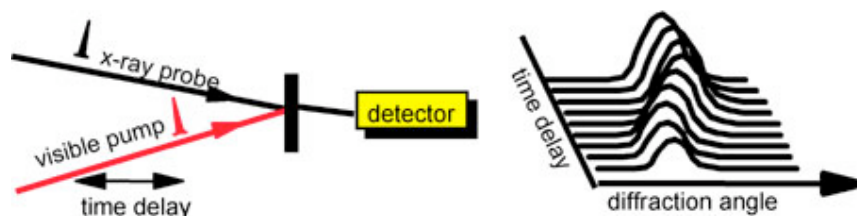
Ultrafast x-ray science

Time-resolved EXAFS, NEXAFS, surface EXAFS

LUX website



Time-resolved x-ray diffraction

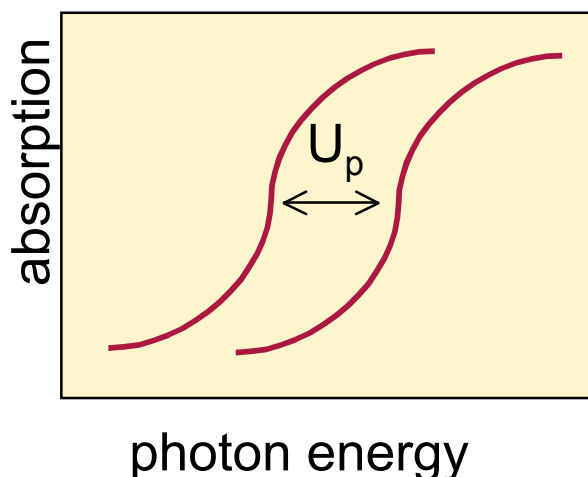


*Laser-pump/x-ray probe techniques central
Short pulses automatically yield high intensities
 $1 \text{ mJ}/100 \text{ fs}/(0.1 \text{ mm})^2 \approx 10^{14} \text{ W/cm}^2 \approx 3 \text{ V/\AA}$*

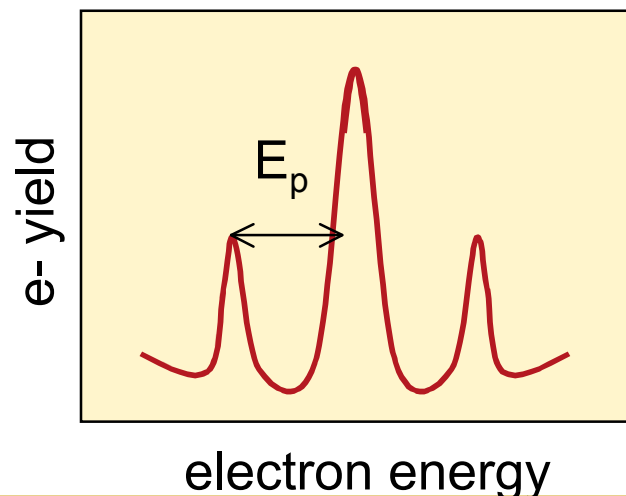
Ultrafast laser/x-ray interactions: isolated atoms

- *X-ray photoionization is fairly well understood in the weak-field limit*
- *Understand changes to x-ray processes in presence of strong laser fields*
- *Theoretical predictions*
 - ponderomotive shift in threshold -> absorption spectrum*
 - free-free transitions in continuum -> electron spectra*

Ponderomotive shift



Electron satellites



Ponderomotive Shift: photodetachment in Cl⁻

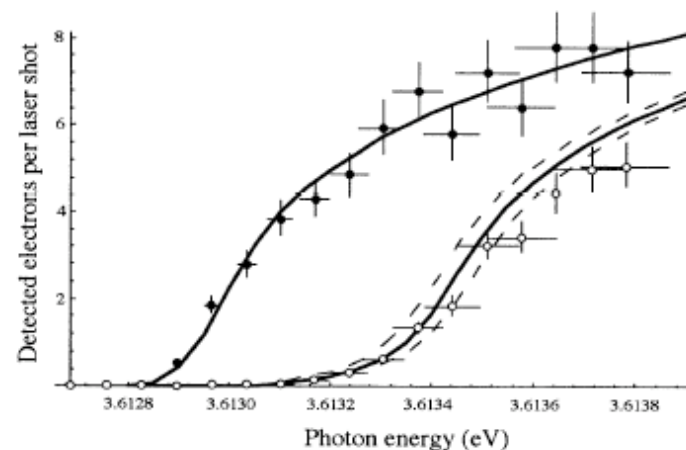
Shift threshold by U_p : $U_p = e^2 E^2 / 4m \omega^2$

$$U_p \approx (9.33 \times 10^{-14} \text{ eV}) I(\text{W/cm}^2) \lambda^2(\mu\text{m})^2$$

M.D. Davidson, J. Wals, H.G. Muller, H.B. van Linden van den Heuvell
PRL 71, 2192 (1993)

IR: 15 mJ, 13 ns, 146 μm
 $I_{1064} = 4.5 \times 10^9 \text{ W/cm}^2$
 $U_p = 0.00045 \text{ eV}$

UV: 14 μJ , 3.4 ns, 36 μm



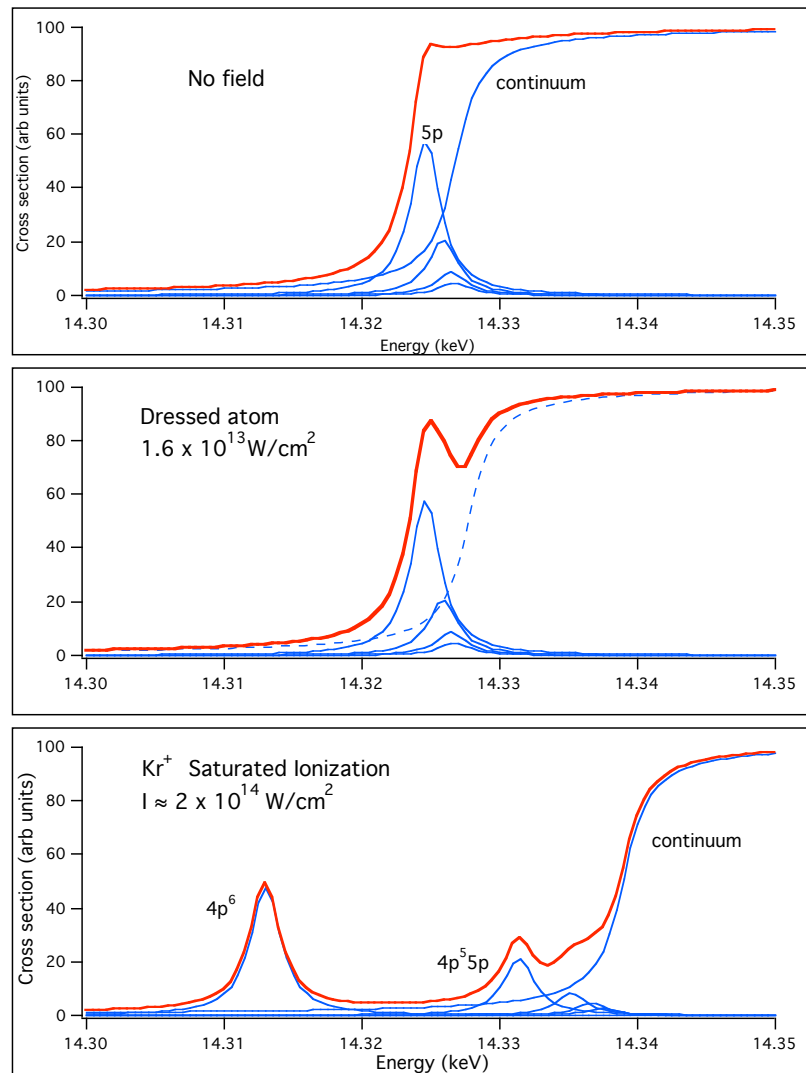
Overlap is critical !!

Well-defined optical field \square dressing beam $\square \gg$ probe \square
 10^{14} W/cm^2 @ 800 nm $\square U_p = 6 \text{ eV}$

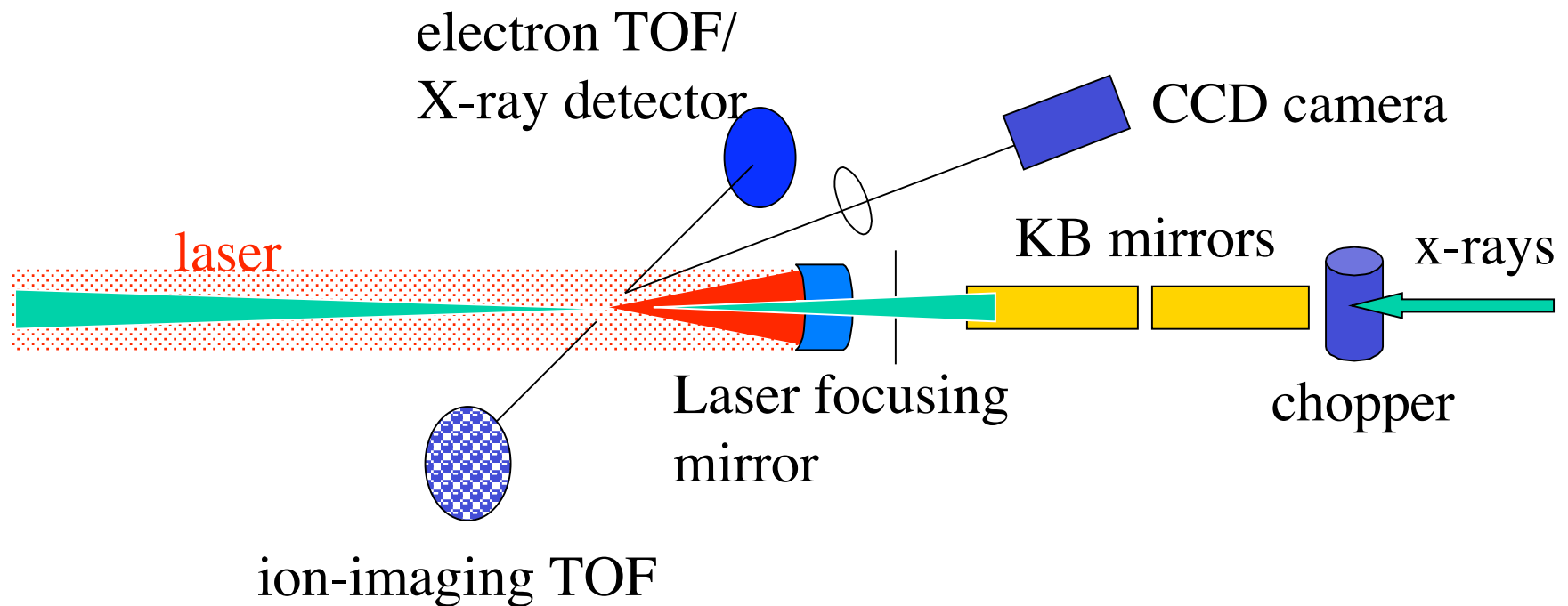
Evolution of Kr 1s-edge structure

- Naked atom: $I=0$
- Dressed atom: $I < I_{\text{sat}}$
- Ion spectrum: $I > I_{\text{sat}}$

For Kr: 800 nm
 $I_{\text{sat}} \approx 2 \times 10^{14} \text{ W/cm}^2$



X-ray processes in presence of strong fields



Technical Challenges

- *Laser intensity*

MHATT-CAT Ti:sapphire laser

$\approx 800 \pm 10$ nm, ≈ 100 fs, ≈ 1 mJ/pulse

To achieve 10^{14} W/cm²

Short pulse (100 fs) focus to ≈ 100 μ m

Long pulse (100 ps) focus to ≈ 3 μ m

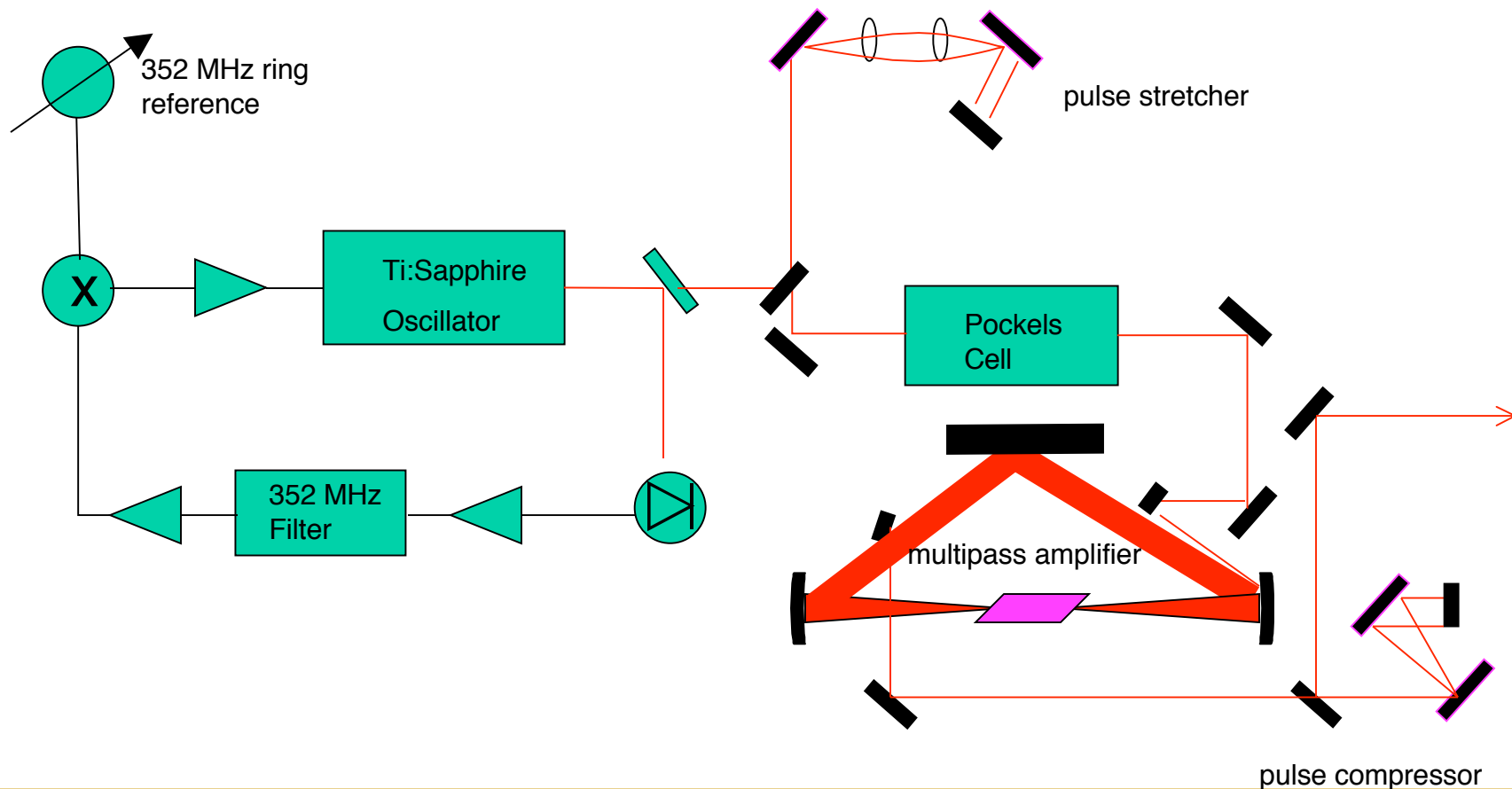
- *Overlap - spatial and temporal*

X-rays: 87 ps, 2 x 3 μ m

- *Count rates*

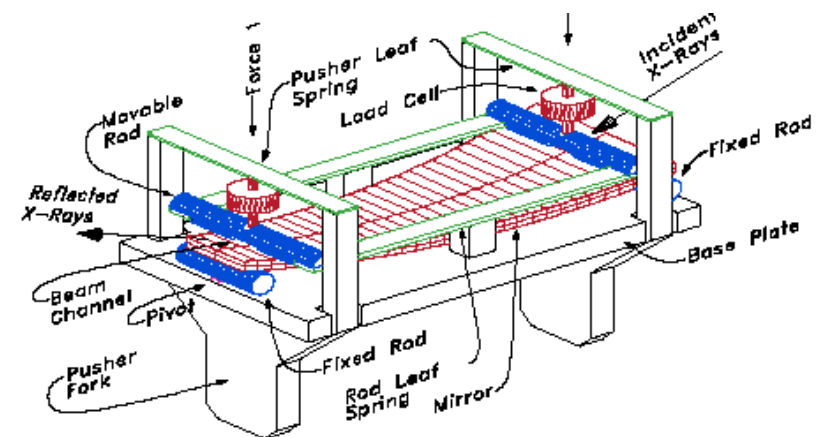
MHATT-CAT Ti:Sapphire Laser System

Oscillator: 88 MHz, 1nJ, 50 fs
Amplifier: 1 kHz, 1mJ, 50 fs



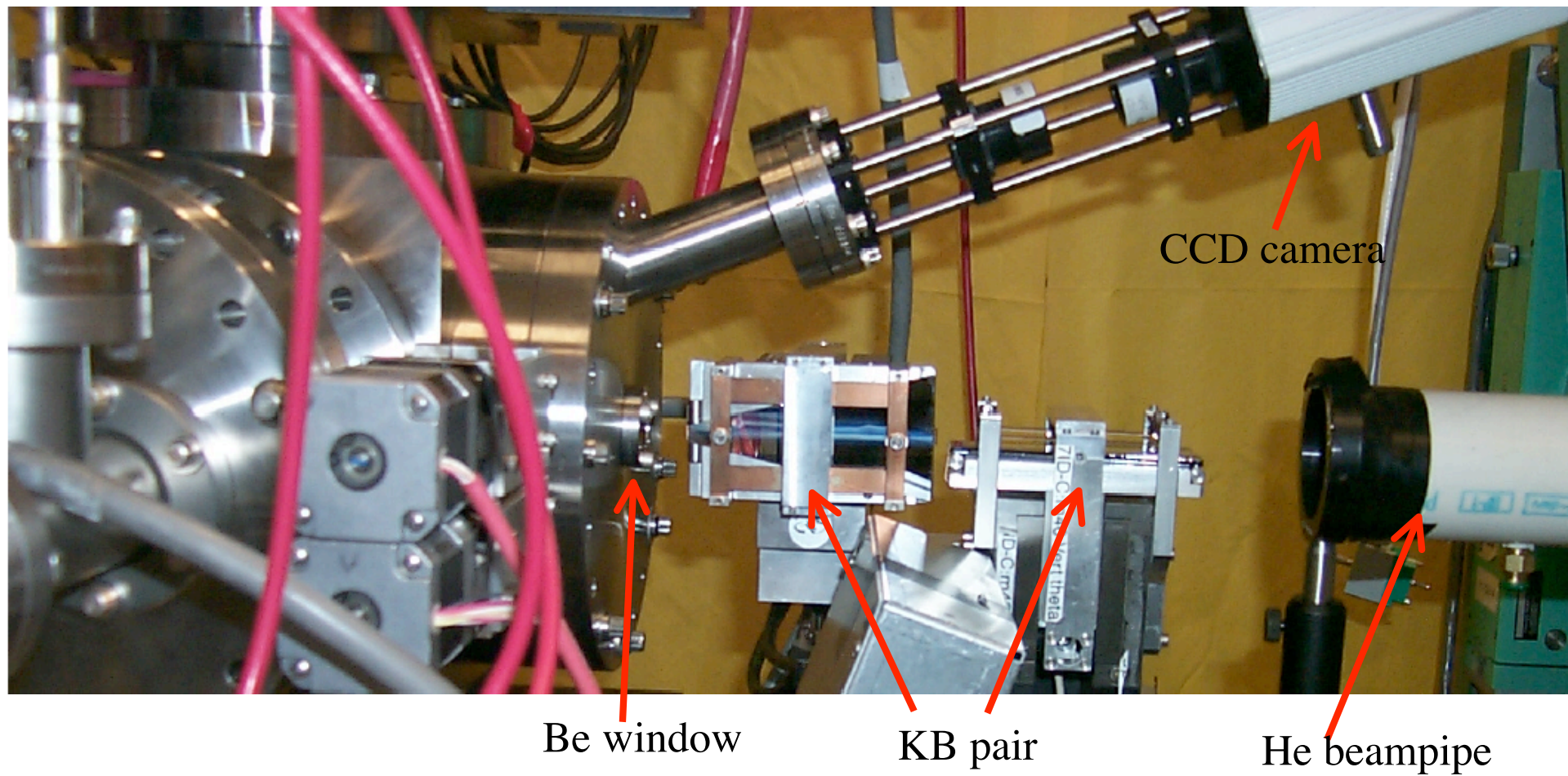
Focus x rays with Kirkpatrick-Baez mirror pair

- *four point bender and trapezoidal mirror substrate is used to create an elliptical shape*
- *a pair is used for vertical and horizontal focusing*
- *focused spot size of $\approx 1 \times 1 \mu\text{m}$ is possible with short working distances*
- *“optimal” spot size $2 \times 3 \mu\text{m}$ calculated for our working distance*



Design by Peter Eng GeoCARS, U Chicago

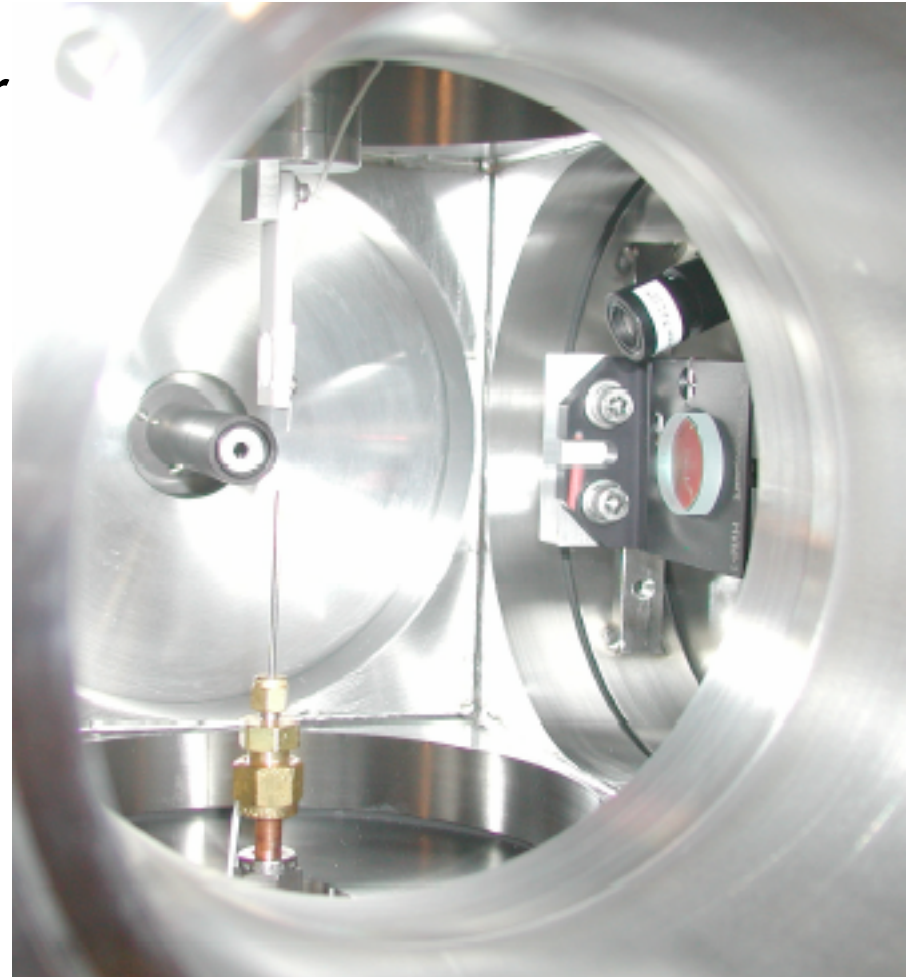
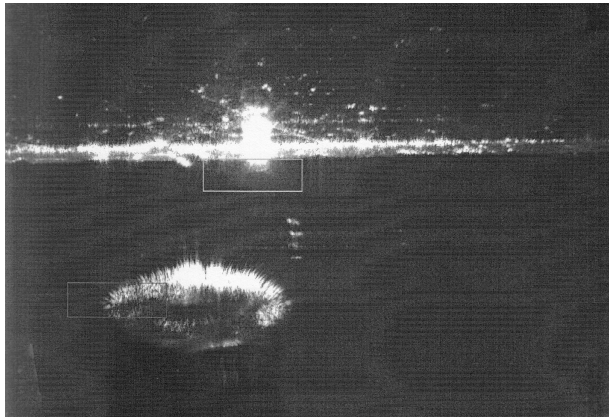
KBs in action



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Alignment Strategy

- ***Focus x rays to center of chamber***
- ***Locate x-ray centroid with BGO***
- ***Overlap focused laser***
 - rough: BGO crystal
 - fine: in-vacuum cross hairs

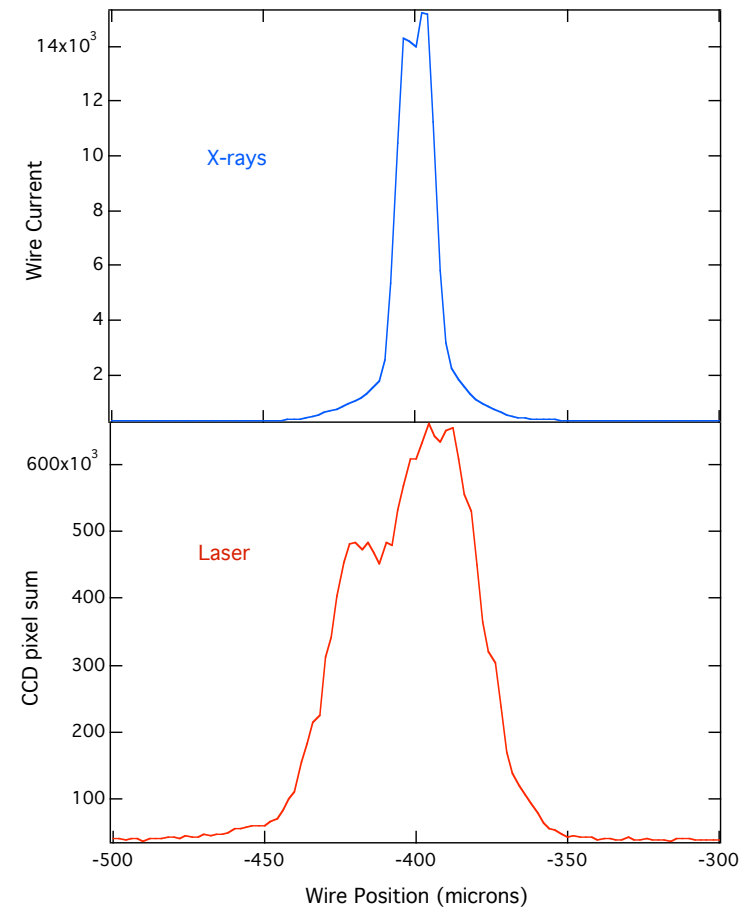


Simultaneous overlap of laser & x-rays

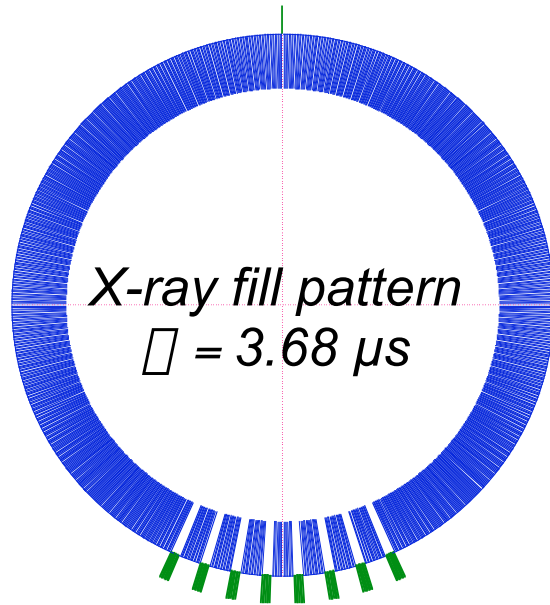
Scan 10 μm cross-hair

X-rays: monitor current
from electrically isolated
cross-hair

Laser: monitor scattered light
pixel sum from a selected
region on CCD camera

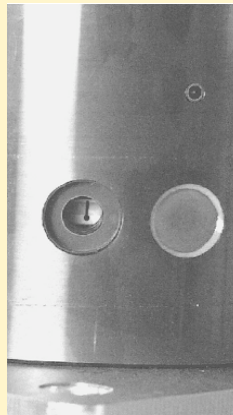


X-ray flux coincident with 1 kHz laser



*Only 1/5440 of x-ray flux is coincident with laser
(overlap with singlet (1/20 total flux) @ 272 kHz)*

*Chopper selects singlet x-ray pulses @ 2.66 kHz
Laser @ 887 Hz : 1 laser-on vs 2 laser-off*



*0.51 mm slot
50.8 mm diameter
2.45 μs open time*

Count rates in Kr atomic beam

Monochromatic x-rays @ 14.3 keV:
 $\approx 10^{13}/s$ ($2 \times 10^6/\text{pulse}$)

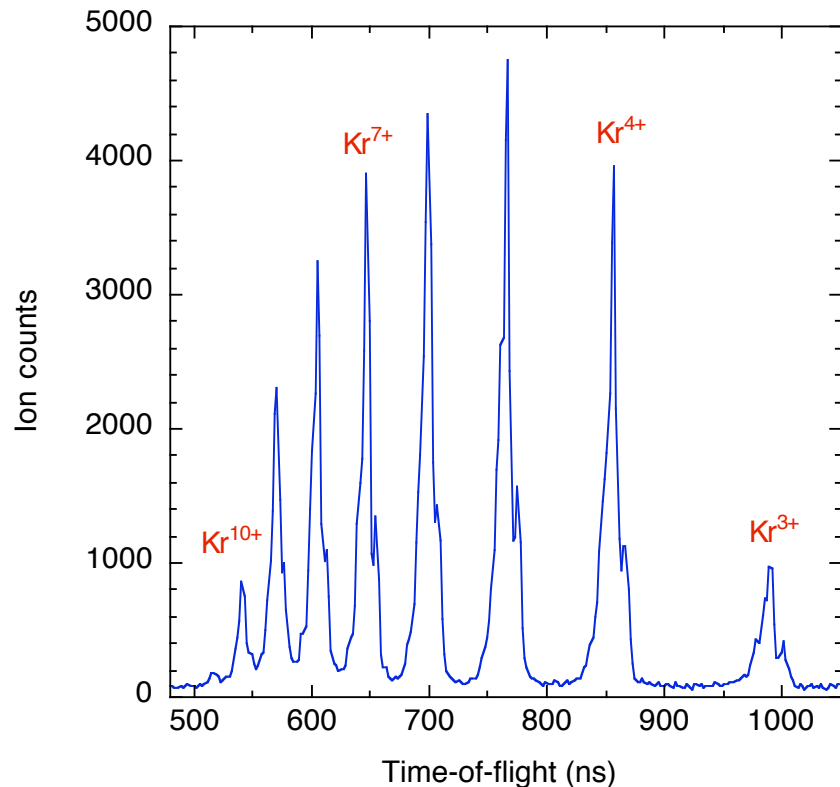
Focused x-rays $\approx 10^{12}/s$

Laser frequency ≈ 887 Hz

$I_{x\text{-ray}}(\text{laser-on}) \approx 2 \times 10^8/s$

$\square = 18$ kb, $n \approx 10^{12} \text{ cm}^{-3}$, $L \approx 0.3 \text{ cm}$

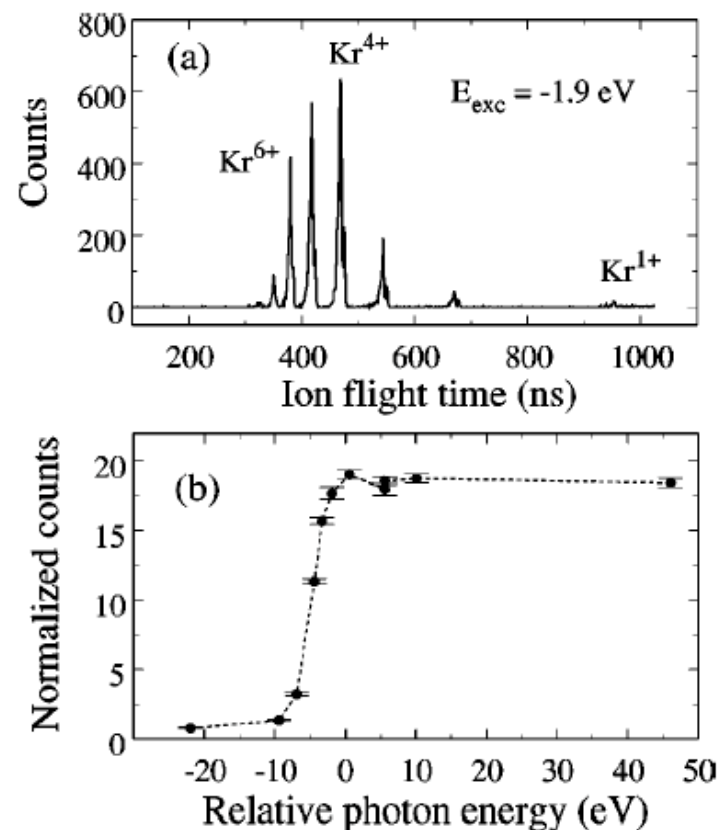
Rate $\approx I \square n L \approx 1$ Hz



Measurement of Kr 1s Near Edge Spectrum

- Ion or x-ray yield to measure absorption spectrum
- Measure threshold structure point by point
- Cross-correlation: laser + x-rays via ion charge state spectroscopy

**Spectrum: 50 points x 10 min/point
500 minutes**

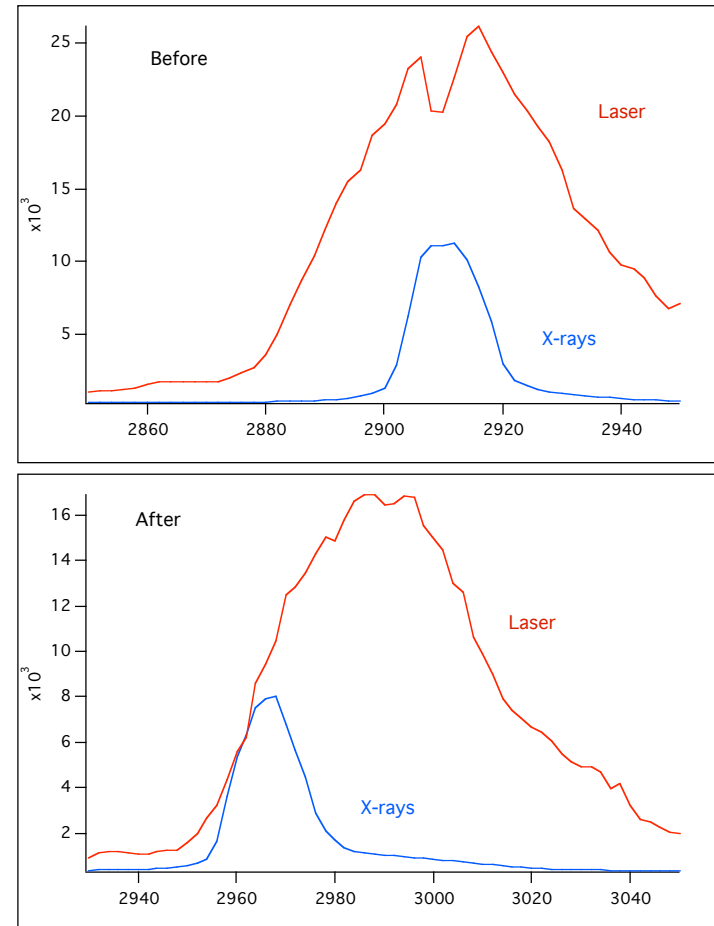


“Stability” of laser/x-ray overlap

*Spatial overlap drifts ≈ 30 microns/4 hrs
Beam position stabilization required*

Temporal overlap was stable

Vertical Wire Scan



Summary and outlook

- *Goal: Understand changes to x-ray processes in presence of strong-laser fields*
- *Technical issues solved*
 - *Overlap*
 - *Dressing intensity (in short pulse mode)*
 - *Count rate*
 - *Stability of overlap*
- *Unique capabilities and personnel associated with APS Sector 7*
- *Next run June 2004*